DC Susceptibility of Interacting Single Domain Particles

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Abstract—The thermal relaxation process in interacting systems is characterized by transitions between metastable configurations of the entire array. Their rates are identified here for sample small particle arrays and the nonequilibrium initial dc susceptibility $\chi(t)$ is found from a master equation. The function $\chi(t)$ has multiexponential time dependence and may exhibit nonuniform approach to equilibrium.

Master equation formalism [1], [2] is applied here to calculations of initial dc susceptibility of a small number of identical magnetostatically coupled uniaxial particles. Assuming coherent rotation we write their energy as $E = KV \sin^2 \theta - V \vec{M} \cdot \vec{H}$ where V is the activation volume, K the anisotropy constant, $\vec{M}$ the magnetization vector ($|\vec{M}| = M_s$) and $\vec{H}$ is the applied field. The angle θ is spanned by the magnetization vector $\vec{M}$ and the easy axis of the particle. The energy of magnetostatic coupling between a pair of particles at a distance $\vec{r}$ from each other is $V^{-2} E_{\text{int}} = r^{-3}(\vec{M}_1 \cdot \vec{M}_2) - 3r^{-5}(\vec{M}_1 \cdot \vec{r})(\vec{M}_2 \cdot \vec{r})$ and $r = |\vec{r}|$. We introduce yet the nucleation field $H_n = 2K/M_s$, the reduced barrier height $q = KV/(k_B T)$ where T is temperature, and, following Chen *et al.* [3], also the dimensionless coupling constant $\rho = M_s^2 V / (2K r^3)$.

The master equation formalism is introduced as follows: Let $n_i(t)$ be the probability that the i th metastable state of a thermally relaxing system is occupied at time t . Then

$$\dot{n}_i = \sum_{j=1}^{\mathcal{N}} R_{ij} n_j \quad \text{with} \quad \sum_{i=1}^{\mathcal{N}} n_i(t) = 1 \quad (1)$$

where $\dot{n}_i = dn_i/dt$, $\mathcal{N}$ is the number of available metastable states and R_{ij} is the rate of the transition $i \rightarrow j$. The elements R_{ij} are well known for noninteracting particles [4] ($\mathcal{N} = 2$) and recently [1]–[3] they were also derived for coupled pairs of parallel particles with bond angles $\beta = 0$ and $\pi/2$ ($\mathcal{N} = 3$). To the first order in small coupling strength ρ we found [2] the matrix R also for $N > 2$ particles. If the system is in equilibrium in zero field then the equilibrium probabilities satisfy $\sum_j R_{ij}(0, T) n_{j0} = 0$ and the system's response to an infinitesimal perturbing field δH is found by substituting the expansions $n_i \approx n_{i0} + \nu_i \delta H$ and $R \approx R(0) + R'(0) \delta H$, $R' = \partial R / \partial H$, into Eq.(1) and isolating the first order terms:

$$\dot{\nu}_i = \sum_{j=1}^{\mathcal{N}} (R_{ij} \nu_j + R'_{ij} n_{j0}) \quad \text{with} \quad \sum_{i=1}^{\mathcal{N}} \nu_i(t) = 0 \quad (2)$$

and initial conditions $\nu_i(0) = 0$. In order to avoid complicated expressions [4] we shall assume that all particles are parallel to the perturbing field so that the magnetization vectors $\vec{M}^{(i)}$, $|\vec{M}^{(i)}| = M^{(i)} \leq M_s$, corresponding to the $\mathcal{N}$ metastable configurations remain unperturbed. The longitudinal initial dc susceptibility $\chi(t)$ then takes on the simple form

$$\frac{H_n}{M_s} \chi(t) = \frac{1}{M_s} \sum_{j=1}^{\mathcal{N}} M^{(j)} \nu_j(t) \stackrel{\text{def}}{=} \hat{\chi}_{\text{eq}}(T) g(t, T) \quad (3)$$

with the equilibrium value $\hat{\chi}_{\text{eq}} = \langle M^2 \rangle / k_B T$. There is, further, $g(0, T) = 0$ and $\lim_{t \rightarrow \infty} g(t, T) = 1$ where the system reaches its perturbed equilibrium. The function $g(t, T)$ has the form

$$g(t) = 1 - \sum_{i=1}^{\mathcal{N}'} g_i(T, \rho) e^{-\kappa_i(T, \rho) t} \quad (4)$$

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with $\sum_{i=1}^{N'} g_i = 1$ and $N' \leq (N+1)/2$ by symmetry of the demagnetized state. The rates $\kappa_{i+1} > \kappa_i > 0$ decrease with temperature according to $\ln \kappa_i \sim -q$.

For noninteracting particles one obtains¹ $\hat{\chi}_{\text{eq}} = 2q$ with $N' = 1$, $g_1 = 1$ and $\kappa_1 = 2f_0 e^{-q}$ in Eq.(4). Analysis [1] of a two particle cluster [3] leads to $\hat{\chi}_{\text{eq}} = 4q/[1 + \varepsilon^2]$ with $N' = 1$, $g_1 = 1$ and $\kappa_1 = 2f_0 \exp[-q(1 + \rho)(1 \pm 3\rho)]$ where $\varepsilon = e^{-4q\rho}$ if $\beta = 0$ (ferromagnetic coupling, upper sign) and $\varepsilon = e^{2q\rho}$ if $\beta = \pi/2$ (antiferromagnetic coupling, lower sign). The function $\hat{\chi}_{\text{eq}}(q)$ is monotonous for $\beta = 0$ but, due to the stability of the demagnetized state $\uparrow\downarrow$, it has a local maximum at $4q\rho \approx 1.28$ if $\beta = \pi/2$.

The weak coupling results, $q\rho \ll 1$, are comparable to a mean field theory with $H \rightarrow H + \alpha M(t)/M_s$ where $M(t)$ is the instantaneous magnetization and α an empirical coupling constant: $\hat{\chi}_{\text{eq}} = 2q/\bar{\alpha}$ and $\kappa_1 = 2f_0 e^{-q\bar{\alpha}}$ where $\bar{\alpha} = 1 - 2q\alpha$ and $|q\alpha| \ll 1$ [4].

In order to study a multiexponential process we consider next a chain of four weakly coupled, equidistant particles parallel to δH with nearest neighbor interactions. There is now $N = 10$ and $N' = 4$ and a schematic representation of the matrix R is:

where $\bullet$ = "spin up" and $\circ$ = "spin down" states and solid lines correspond to admissible single particle switching events [2]. There is, further,

$$\hat{\chi}_{\text{eq}} = 4q(2\varepsilon^{-3} + \varepsilon^{-1} + \varepsilon)/[\varepsilon^{-3} + 3(\varepsilon^{-1} + \varepsilon) + \varepsilon^3]$$

with the above notation, and the properties of the function $g(t, T)$ are summarized in Fig.1: Simple exponential decay is observed only if $q\rho \rightarrow 0$, in this case $g_1 \rightarrow 1$ and $g_i \rightarrow 0$ for $i \geq 2$. If, on the other hand, $q\rho \rightarrow \infty$ then $\kappa_1 \ll \kappa_2$ for either value of the bond angle β , but, the coefficients g_i , $i \geq 2$, become

¹In calculations we take the prefactor as $f_0 = e^{25}$ Hz.

large, influencing the initial response (Fig.1). The unexpected feature of nonuniform approach to equilibrium is encountered in the transition region $4q\rho \approx 1$ for $\beta = \pi/2$ where $\hat{\chi}_{\text{eq}}$ has a local maximum: At time $t = t_0 \sim \kappa_1^{-1}$ the function $g(t)$ has a local maximum $g(t_0) > 1$ as well and henceforth it decreases towards the stationary value $\lim_{t \rightarrow \infty} g(t) = 1$, the difference $g(t_0) - 1 \sim \rho$. This suggests that the result of a finite time measurement may exceed the equilibrium value $\hat{\chi}_{\text{eq}}(T, \rho)$. Similar behavior was encountered also in a chain of six particles with periodic boundary conditions ($N = 13$, $N' = 6$). Unfortunately, no solvable multiexponential model valid for all ρ is known to us and the dependence of the excess value $g(t_0) - 1$ on large ρ could not be studied.

Our last model is a planar array of weakly coupled particles [2] deposited on a (possibly stretched) square lattice, at $(ri, 0, r\xi j)$, $i, j \in (-\infty, \infty)$, $\xi > 0$ is the stretching factor and r is now a lattice constant. All particles are parallel to δH applied in the stretched direction and the model approximates the lithographically deposited lattices of J. F. Smythe *et al.* [5]. In each metastable configuration the entire

Fig.1. The function $g(t, q)$ for a four particle chain versus the scaled time $\kappa_1 t$. Dashed line: single exponential decay at $\rho = 0$. Solid lines above this curve correspond to $\rho = 0.015$ and $\beta = \pi/2$ (relaxation is accelerated) and below to $\rho = 0.005$ and $\beta = 0$ (relaxation is slowed down). These curves successively deviate from the decoupled result with increasing inverse temperature $q \in (2, 42)$. The $\beta = \pi/2$ curves slightly overshoot unity for coupling strengths $4q\rho \approx 1$.

Fig. 2. The equilibrium and nonequilibrium reduced susceptibilities $\hat{\chi}_{eq}$ (dashed lines) and $\hat{\chi}_{eq}g(t, T)$ (solid lines) versus inverse temperature q . Stretching factor successively $\xi = 0.65$ (as shown), 0.75, 1 (the highest peak), 1.25, 1.35, 1.43, 1.5, 1.75 and 2 (as shown). Increased coupling strength (smaller lattice constant) enhances susceptibility at $\xi \approx 1$ but suppresses it for other stretching factors. Waiting time $t = 10$ s in all cases.

plane is assumed to be covered by identical copies of a given metastable configuration of a four particle square cluster and the *long range* interactions are calculated as lattice sums [2], [6]. There is then $\mathcal{N} = 7$ and $\mathcal{N}' = 2$ and the dc susceptibility (3), whose sample plots are shown in Fig. 2, may be written in closed form. The matrix R is in this case a

function of eight independent *single particle* relaxation rates. At large stretching factor ξ the ferromagnetic coupling in the $\vec{k}$ -direction of the plane is suppressed and the system is dominated by antiferromagnetic interactions which decrease the value of $\hat{\chi}_{eq}$ but accelerate the approach of $g(t, T)$ to unity. With decreasing ξ the ferromagnetic component of the magnetostatic field grows and its growth is accompanied by an increase in $\hat{\chi}_{eq}$, a decrease in relaxation rate and, also, by decreasing occurrence probabilities of partially demagnetized metastable configurations [2] till at $\xi < 1$ effectively $\mathcal{N} = 3$ and $\mathcal{N}' = 1$. The two competing processes give rise to the prominent susceptibility peaks at $\xi \approx 1$ in Fig. 2. This transition configuration also exhibits [2] a local maximum of the switching field distribution at coercivity and must therefore be regarded as the least stable against small perturbations.

An alternate venue to studies of thermal relaxation in small particles is offered by the Preisach model [7]. Within this formalism, however, it would probably be difficult to implement the interplay of ferromagnetic and antiferromagnetic coupling and, above all, the effects associated with the changing number of contributing configurations.

REFERENCES

- [1] I. Klik, J. S. Yang and C. R. Chang, "Hysteresis of binary clusters," *J. Appl. Phys.*, vol. 76, pp. 6493-6495, November 1994.
- [2] I. Klik and C. R. Chang, "Thermal relaxation in arrays of coupled ferromagnetic particles," submitted to *Phys. Rev. B*.
- [3] W. Chen, S. Zhang and H. N. Bertram, "Energy barriers for thermal reversal of interacting single domain particles," *J. Appl. Phys.*, vol. 71, pp. 5579-5584, June 1992.
- [4] J. S. Yang, J. Lee, I. Klik and C. R. Chang, "Measuring dc susceptibility of particulate media," *J. Magn. Mat.*, vol. 140-144, pp. 407-408, February 1995.
- [5] J. F. Smythe, S. Schultz, D. R. Fredkin, D. P. Kern, S. A. Rishton, H. Schmid, M. Cali and T. R. Koehler, "Hysteresis of lithographic arrays of permalloy particles: Experiment and theory," *J. Appl. Phys.*, vol. 69, pp. 5262-5266, April 1991.
- [6] J. Lam, "Magnetic hysteresis of a rectangular lattice of interacting single-domain particles," *J. Appl. Phys.*, vol. 72, pp. 5792-5798, December 1992.
- [7] I. D. Mayergoyz and C. E. Korman, "The Preisach model with stochastic input as a model for aftereffect," *J. Appl. Phys.*, vol. 75, pp. 5478-5480, May 1994.